

Deformable dielectrics II Voigt's intramolecular force balance in elastic dielectrics

G. A. MAUGIN (PARIS)

THE FIELD equations developed in the first part of this work for a dynamical theory of non-magnetized deformable dielectrics made of several molecular species are here supplemented with the relevant thermodynamics. This allows for the construction of constitutive equations for thermoelastic solids with finite deformations and the comparison of several previous formulations. As an approximation within the infinitesimal strain theory, the equations that generalize Voigt's intramolecular force balance to the case of elastic ferroelectrics, antiferroelectrics or ferrielectrics are deduced. All thermo-electro-mechanical couplings and the associated loss mechanisms are shown to participate in such equations which must be considered in the study of electroacoustic waves and electro-optical effects.

Wyprowadzone w pierwszej części tej pracy równania polowe dynamicznej teorii dielektryków odkształcalnych składających się z różnych cząsteczek uzupełniają się tutaj odpowiednią termodynamiką. Pozwala to na zbudowanie równań konstytutywnych dla ciał termosprężystych o odkształceniach skończonych oraz na analizę porównawczą kilku wcześniejszych sformułowań. Posługując się przybliżeniem odpowiadającym teorii odkształceń nieskończonych wyprowadzono równania uogólniające warunki równowagi międzycząsteczkowych sił Voigta na przypadek sprężystych ferro-elektryków, anti-ferroelektryków oraz ferri-elektryków. Wykazano, że wszelkie sprzężenia termo-elektromechaniczne oraz odpowiednie mechanizmy strat występują w tych równaniach i należy je uwzględnić przy rozważaniu fal elektroakustycznych i zjawisk elektro-optycznych.

Выведенные в первой части этой работы полевые уравнения динамической теории деформируемых диэлектриков, состоящих из разных молекул, пополняются здесь соответственной термодинамикой. Это позволяет построить определяющие уравнение для термоупругих тел с конечными деформациями, а также провести сравнительный анализ ряда раньше полученных формулировок. Послуживаясь приближением, отвечающим теории инфинитезимальных деформаций, выведены уравнения обобщающие условия равновесия межмолекулярных сил Фойхта на случай упругих ферроэлектриков, анти-ферроэлектриков и ферриэлектриков. Показано, что всякие термо-электромеханические сопряжения и соответственные механизмы потерь выступают в этих уравнениях и следует их учитывать при рассмотрении электроакустических волн и электро-оптических явлений.

1. Introduction

THE PRESENT paper is mostly devoted to constructing the constitutive equations for the dynamical theory of nonmagnetized deformable dielectrics made of several molecular species set forth in Part One. To this purpose the local statements of the energy equation and of the Clausius-Duhem inequality are first derived (Section 3) after having recalled the essential results of Part One (Section 2). Several forms of the Clausius-Duhem inequality are given which prove appropriate depending on the choice of dependent variables. Then, using the thermodynamical admissibility of Coleman, we deduce two alternate formulations of the constitutive equations for a thermoelastic solid subject to finite deformations (Section 4). A special attention is devoted to the infinitesimal strain theory (expansion of the free

energy about a strain-free state of spatially uniform temperature) in order to exhibit the different effects accounted for: thermoelasticity, pyroelectricity, piezoelectricity, electrostriction, and the interactions between the different polarization sub-lattices. Kittel's theory of rigid antiferroelectric is thus generalized to the case of anisotropic thermoelastic bodies. Voigt's theory of piezoelectricity also appears to be a very peculiar case. Taking account of thermodynamically irreversible processes in agreement with the linear Onsager-Casimir theory for the sake of example, it is then possible (in Section 5) to show, owing to the decomposition of the Cauchy stress obtained in Part One, that the dielectric relaxation term in general contributes, along with the usual viscosity processes, to the stress tensor in a ferroelectric, which indicates at once that the damping of elastic waves and polarons (oscillations in the electric moments) will be coupled. It is finally argued that, on account of the polarization inertia, of the dielectric loss mechanisms and of the deformation field, the intramolecular force balance derived for each polarization sub-lattice generalizes the early proposal of Voigt, so as to allow for a thorough analysis of electro-acoustic waves and electro-optical effects for various states of dielectricity in an elastic dielectric.

2. Summary of previous results

In a previous paper [1] referred to as Part One herein after^(*), the following field equations were obtained for a moving nonmagnetized deformable dielectric made of several molecular species (in a spatial region \mathcal{D}_t boundary by $\partial\mathcal{D}_t$):

a. *Maxwell's equations in \mathcal{D}_t* (using Lorentz-Heaviside units)

$$(2.1) \quad \begin{aligned} \nabla \times \mathbf{E} + \frac{1}{c} \frac{\partial \mathbf{B}}{\partial t} &= \mathbf{0}, & \nabla \cdot \mathbf{B} &= 0, \\ \nabla \times \mathbf{B} - \frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} &= \frac{1}{c} [\dot{\mathbf{P}} - (\mathbf{V} \cdot \mathbf{P})\mathbf{U}], & \nabla \cdot \mathbf{E} &= -\nabla \cdot \mathbf{P}. \end{aligned}$$

b. *Continuity equation in \mathcal{D}_t*

$$(2.2) \quad \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{U}) = 0 \quad \text{or} \quad \dot{\rho} + \rho \nabla \cdot \mathbf{U} = 0.$$

c. *Motion equation*

$$(2.3) \quad \rho \dot{\mathbf{U}} = \text{div } \mathbf{t} + \mathbf{f} + \frac{1}{c} \dot{\mathbf{P}} \times \mathbf{B} + (\mathbf{P} \cdot \nabla) \mathcal{E} \quad \text{in } \mathcal{D}_t,$$

$$(2.4) \quad T_i + T_i^{em} = t_{ij} n_j \quad \text{on } \partial\mathcal{D}_t,$$

with

$$(2.5) \quad \mathcal{E} \equiv \mathbf{E} + \frac{1}{c} \mathbf{U} \times \mathbf{B},$$

$$(2.6) \quad t_{ij} = \sigma_{ij} + \rho \sum_{\alpha} c_{\alpha}^L E_{\alpha[i} \pi_{\alpha j]},$$

^(*) Equations of Part One are referred to by I followed by their number, e.g., Eq. (I-2.18). We refer to Part I for the main notation.

$$(2.7) \quad T_i^{em} = - \left[\delta_i P_j + E_i E_j + B_i B_j - \frac{1}{2} (\mathbf{E}^2 + \mathbf{B}^2) \delta_{ij} \right] n_j,$$

$$(2.8) \quad \mathbf{P} = \varrho \sum_{\alpha} c_{\alpha} \boldsymbol{\pi}_{\alpha}, \quad \mathbf{P}_{\alpha} = \varrho_{\alpha} \boldsymbol{\pi}_{\alpha}.$$

d. *Intramolecular force balance laws;*

$$(2.9) \quad \mathcal{E} + {}^L \mathbf{E}_{\alpha} = d_{\alpha} \dot{\boldsymbol{\pi}}_{\alpha}, \quad \alpha = 1, 2, \dots, n, \quad \text{in } \mathcal{D}_t.$$

We recall that c_{α} is the time-constant concentration in α -molecular species, the latter giving rise to an electric polarization field $\boldsymbol{\pi}_{\alpha}$ per unit mass of α species. d_{α} is the so-called molecular polarization inertia for the α species. $\dot{\mathbf{P}}$ is the convected time-derivative of the total volume polarization \mathbf{P} defined as

$$\dot{\mathbf{P}} = \mathbf{P} - (\mathbf{P} \cdot \nabla) \mathbf{U} + \mathbf{P} (\nabla \cdot \mathbf{U}).$$

To close the differential system formed by Eqs. (2.1), (2.2), (2.3) and (2.9), constitutive equations must be constructed for the symmetric "intrinsic stress" tensor σ_{ij} and for the "local electric" fields ${}^L \mathbf{E}_{\alpha}$, $\alpha = 1, 2, \dots, n$. The following global energy expression which was established on the basis of the expression of the principle of virtual power and of an energy identity satisfied by the electromagnetic fields (cf. Eq. I-4.31), proves to be essential in this context:

$$(2.10) \quad \dot{K}(\mathcal{D}_t) + \dot{U}^{em}(\mathcal{D}_t) = \mathcal{P}_{(i)}(\mathcal{D}_t) + \int_{\mathcal{D}_t} \mathbf{f} \cdot \mathbf{U} dv + \int_{\partial \mathcal{D}_t} (\mathbf{T} \cdot \mathbf{U} - \mathfrak{J} \cdot \mathbf{n}) da,$$

where

$$(2.11) \quad \begin{aligned} K(\mathcal{D}_t) &= \int_{\mathcal{D}_t} \frac{1}{2} \varrho \left(\mathbf{U}^2 + \sum_{\alpha} c_{\alpha} d_{\alpha} \dot{\boldsymbol{\pi}}_{\alpha}^2 \right) dv, \\ U^{em}(\mathcal{D}_t) &= \int_{\mathcal{D}_t} \frac{1}{2} (\mathbf{E}^2 + \mathbf{B}^2) dv, \\ \mathcal{P}_{(i)}(\mathcal{D}_t) &= - \int_{\mathcal{D}_t} \left(\sigma_{ij} D_{ij} - \varrho \sum_{\alpha} c_{\alpha} {}^L \mathbf{E}_{\alpha} \cdot \hat{\boldsymbol{\pi}}_{\alpha} \right) dv, \end{aligned}$$

in which

$$(2.12) \quad D_{ij} \equiv U_{(i,j)}, \quad (\hat{\boldsymbol{\pi}}_{\alpha})_i \equiv (D_j \boldsymbol{\pi}_{\alpha})_i \equiv \dot{\boldsymbol{\pi}}_{\alpha i} - U_{[i,j]} \boldsymbol{\pi}_{\alpha j},$$

$$(2.13) \quad \mathfrak{J} \equiv c \mathcal{E} \times \left(\mathbf{B} - \frac{1}{c} \mathbf{U} \times \mathbf{E} \right),$$

D_j denotes the so-called Jaumann or co-rotational derivative. $\mathcal{P}_{(i)}(\mathcal{D}_t)$ is the power developed by the internal forces in real velocity fields.

3. Thermodynamics

The first and second principles of thermodynamics are postulated in global form independently of the principle of virtual power. They assume the natural forms:

$$(3.1) \quad \frac{d}{dt} [K(\mathcal{D}_t) + E(\mathcal{D}_t) + U^{em}(\mathcal{D}_t)] = \dot{Q}(\bar{\mathcal{D}}_t) + \int_{\mathcal{D}_t} \mathbf{f} \cdot \mathbf{U} dv + \int_{\partial \mathcal{D}_t} \mathbf{T} \cdot \mathbf{U} da$$

and

$$(3.2) \quad \dot{N}(\mathcal{D}_t) \geq \dot{\mathcal{S}}(\bar{\mathcal{D}}_t),$$

where

$$(3.3) \quad E(\mathcal{D}_t) = \int_{\mathcal{D}_t} \rho e dv,$$

$$(3.4) \quad \dot{Q}(\bar{\mathcal{D}}_t) = \int_{\mathcal{D}_t} \rho h dv - \int_{\partial \mathcal{D}_t} \mathbf{q} \cdot \mathbf{n} da,$$

$$(3.5) \quad N(\mathcal{D}_t) = \int_{\mathcal{D}_t} \rho \eta dv,$$

$$(3.6) \quad \dot{\mathcal{S}}(\mathcal{D}_t) = \int_{\mathcal{D}_t} \theta^{-1} \rho h dv - \int_{\partial \mathcal{D}_t} \theta^{-1} (\mathbf{q} - \mathfrak{J}) \cdot \mathbf{n} da,$$

in which e , h , \mathbf{q} , η and θ are, respectively, the internal energy per unit mass, the heat (radiation) source per unit mass, the heat influx vector, the entropy per unit mass, and the thermodynamical temperature ($\theta > 0$, $\inf \theta = 0$). We followed ERINGEN [2] in postulating the entropy influx as $\theta^{-1} \tilde{\mathbf{q}}$, where

$$(3.7) \quad \tilde{\mathbf{q}} \equiv \mathbf{q} - \mathfrak{J};$$

that is θ^{-1} is assumed to be an integrating factor and the E. M. energy flux participates in the entropy influx. Such a formulation can be *derived* by using Müller's thermodynamics (See LIU and MÜLLER [3]).

Combining Eqs. (3.1) and (2.10), we obtain the global statement of the *theorem of the energy*:

$$(3.8) \quad \dot{E}(\mathcal{D}_t) + \mathcal{P}_{(t)}(\mathcal{D}_t) = \int_{\mathcal{D}_t} \rho h dv - \int_{\partial \mathcal{D}_t} \tilde{\mathbf{q}} \cdot \mathbf{n} da,$$

whose local form reads

$$(3.9) \quad \rho \dot{e} = \sigma_{ij} D_{ij} - \rho \sum_{\alpha} c_{\alpha}^L \mathbf{E}_{\alpha} \cdot \hat{\mathbf{\Pi}}_{\alpha} - \nabla \cdot \tilde{\mathbf{q}} + \rho h.$$

Combining the local form of Eq. (3.2) with Eq. (3.9), so as to eliminate h , we are led to the *Clausius-Duhem inequality* in the form

$$(3.10) \quad -\rho(\dot{\psi} + \eta\dot{\theta}) + \sigma_{ij} D_{ij} - \rho \sum_{\alpha} c_{\alpha}^L \mathbf{E}_{\alpha} \cdot \hat{\mathbf{\Pi}}_{\alpha} - \theta^{-1} \tilde{\mathbf{q}} \cdot \nabla \theta \geq 0,$$

where $\psi = e - \eta\theta$ is the free energy per unit mass. Other forms can be given to this inequality. First, using the definition (2.12)₂ and taking account of Eq. (2.6), Eq. (3.10) is transformed to

$$(3.11) \quad -\rho(\dot{\psi} + \eta\dot{\theta}) + t_{ij} U_{i,j} - \rho \sum_{\alpha} c_{\alpha}^L \mathbf{E}_{\alpha} \cdot \dot{\mathbf{\Pi}}_{\alpha} - \theta^{-1} \tilde{\mathbf{q}} \cdot \nabla \theta \geq 0.$$

Next, upon using the demonstrable result (cf. Eq. 1-3.7)

$$(3.12) \quad \rho_{\alpha} \hat{\mathbf{\Pi}}_{\alpha i} = \dot{P}_{\alpha i} + \rho_{\alpha} D_{ij} \pi_{\alpha j},$$

where $\dot{\mathbf{P}}_\alpha$ is the convected time-derivative of \mathbf{P}_α —remark that $\dot{c}_\alpha = 0$ —Eq. (3.10) transforms to

$$(3.13) \quad -\varrho(\dot{\psi} + \eta\dot{\theta}) + {}^E t_{ij} D_{ij} - \sum_\alpha {}^L \mathbf{E}_\alpha \cdot \dot{\mathbf{P}}_\alpha - \theta^{-1} \tilde{\mathbf{q}} \cdot \nabla\theta \geq 0,$$

where

$$(3.14) \quad {}^E t_{ij} \equiv \sigma_{ij} - \varrho \sum_\alpha c_\alpha {}^L E_{\alpha(i}\pi_{\alpha j)} = {}^E t_{ji}$$

will be referred to as the (symmetric) “elastic” stress tensor.

The following remarks pertain to the different forms (3.10), (3.11) and (3.13). On the one hand, Eq. (3.11) makes clear the contribution of the Cauchy stress to the Clausius-Duhem inequality, while on the other hand it contains nonobjective time rates, so that it should not be used in applying the thermodynamical admissibility of COLEMAN [4]. The inequalities (3.10) and (3.13) contain objective time rates and are thus admissible. However, whereas Eq. (3.10) contains the co-rotational derivatives $\hat{\mathbf{P}}_\alpha$, Eq. (3.13) involves convected-time derivatives $\dot{\mathbf{P}}_\alpha$. It must be noticed in this regard that the use of the first type of derivative is particularly well-adapted whenever the rotational effects are prominent (e.g., in ferromagnetic relaxation in deformable bodies [5]), whereas the convected time-derivative takes account of the general deformation of the medium (including shear and dilatation). But, as was already pointed out in previous papers of ours (e.g., [6]), it is only Eq. (3.10) that contains linearly independent rates, this independence being required for using Coleman’s argument. Thus we shall stick to Eq. (3.10) when using this argument (i.e., in elasticity), but we shall freely use Eq. (3.13) if we call for a different thermodynamic theory such as the Onsager-Casimir theory of irreversible processes.

4. Thermoelastic dielectrics

4.1. General case

Let $x = \mathfrak{X}(X, t)$ be the motion of the deformable dielectric, assumed to be invertible and sufficiently differentiable. Then,

$$(4.1) \quad J = \varrho_0/\varrho = \det \mathbf{F} > 0, \quad F_{iK} \equiv x_{i,K}, \quad F_{Ki}^{-1} \equiv X_{K,i},$$

where ϱ and ϱ_0 are the matter densities in the present and reference configurations, respectively, define the Jacobian of the motion, the deformation gradient, and its reciprocal.

A hyperelastic dielectric is a nonlinear elastic dielectric of which the constitutive equations are derivable from an energy potential, e.g., ψ . In accordance with the working hypothesis of equipresence we assume that all dependent variables ψ , η , σ_{ij} , ${}^L \mathbf{E}_\alpha$ and $\tilde{\mathbf{q}}$ may *a priori* all depend on the same set of independent variable $(\mathbf{F}, \pi_\alpha, \theta, \mathbf{g})$ —where $\mathbf{g} \equiv \nabla\theta$ —evaluated at the present time. For instance,

$$(4.2) \quad \psi = \psi(\mathbf{F}, \pi_\alpha, \theta, \mathbf{g}), \quad \alpha = 1, 2, \dots, n.$$

It is readily shown that for ψ to be *objective* it is sufficient and necessary that it has, for instance, the following reduced functional dependence:

$$(4.3) \quad \psi = \tilde{\psi}(\mathbf{E}, \mathbf{\Pi}_\alpha, \theta, \mathbf{G}),$$

where (T = transpose)

$$(4.4) \quad \mathbf{E} \equiv \frac{1}{2}(\mathbf{F}^T \mathbf{F} - \mathbf{I}), \quad \mathbf{\Pi}_\alpha \equiv \mathbf{F}^T \pi_\alpha, \quad \mathbf{G} \equiv \mathbf{F}^T \mathbf{g}.$$

Using the following auxiliary results

$$(4.5) \quad \dot{E}_{KL} = x_{i,K} D_{ij} x_{j,L}, \quad \dot{I}_{\alpha K} = (\hat{I}_{\alpha i} + D_{ij} \pi_{\alpha j}) x_{i,K},$$

one shows that

$$(4.6) \quad \dot{\psi} = \left(\frac{\partial \tilde{\psi}}{\partial E_{KL}} x_{i,K} x_{j,L} + \sum_\alpha \frac{\partial \tilde{\psi}}{\partial I_{\alpha K}} \pi_{\alpha j} x_{i,K} \right) D_{ij} + \sum_\alpha \frac{\partial \tilde{\psi}}{\partial I_{\alpha K}} x_{i,K} \hat{I}_{\alpha i} + \frac{\partial \tilde{\psi}}{\partial \theta} \dot{\theta} + \frac{\partial \tilde{\psi}}{\partial \mathbf{G}} \cdot \dot{\mathbf{G}}.$$

Substituting from the latter in the inequality (3.10) assumed to be satisfied for arbitrary independent objective time rates (D_{ij} , $\hat{I}_{\alpha i}$, $\dot{\theta}$, $\dot{\mathbf{G}}$), one deduces in the usual fashion the following result:

THEOREM. The constitutive equations of a nonmagnetized hyperelastic dielectric made of several molecular species read

$$(4.7) \quad {}^R \sigma_{ij} = \rho \left(\frac{\partial \hat{\psi}}{\partial E_{KL}} x_{(i,K} + \sum_\alpha \frac{\partial \hat{\psi}}{\partial I_{\alpha L}} \pi_{\alpha(i)} \right) x_{j),L} = {}^R \sigma_{ji},$$

$$(4.8) \quad {}^{RL} E_{\alpha i} = -c_\alpha^{-1} \frac{\partial \hat{\psi}}{\partial I_{\alpha K}} x_{i,K}, \quad \alpha = 1, 2, \dots, n,$$

$$(4.9) \quad \eta = -\frac{\partial \hat{\psi}}{\partial \theta}, \quad \psi = \hat{\psi}(\mathbf{E}, \mathbf{\Pi}_\alpha, \theta),$$

$$(4.10) \quad \tilde{q}_i = x_{i,M} \hat{Q}_M(\mathbf{E}, \mathbf{\Pi}_\alpha, \theta, \mathbf{G}), \quad \hat{\mathbf{Q}} \cdot \mathbf{G} \leq 0,$$

$\hat{Q}_M = X_{M,i} \tilde{q}_i$ being subjected to the "continuity" condition

$$(4.11) \quad \hat{\mathbf{Q}} = \mathbf{0} \quad \text{whenever} \quad \mathbf{G} = \mathbf{0},$$

if it is assumed to be of class C^1 with respect to its argument \mathbf{G} .

The left superscripts R —for (thermodynamically) recoverable—indicate that thus labeled quantities are derived from a thermodynamical potential.

On account of Eqs. (4.7)–(4.8), the thermodynamically recoverable part, ${}^R t_{ij}$, of the Cauchy stress can be written in the form

$$(4.12) \quad {}^R t_{ij} = \rho \left(\frac{\partial \hat{\psi}}{\partial E_{KL}} x_{i,K} + \sum_\alpha \frac{\partial \hat{\psi}}{\partial I_{\alpha L}} \pi_{\alpha i} \right) x_{j,L}$$

or

$$(4.13) \quad {}^R t_{ij} = E_{ij}^{\hat{}} - \rho \sum_\alpha c_\alpha \pi_{\alpha i} {}^{RL} E_{\alpha j},$$

where

$$(4.14) \quad \hat{E}^t_{ij} = \varrho \frac{\partial \hat{\psi}}{\partial E_{KL}} x_{i,K} x_{j,L} = \hat{E}^t_{ji} = {}^R\sigma_{ij} + \varrho \sum_{\alpha} c_{\alpha} {}^{RL}E_{\alpha(i}\pi_{\alpha j)},$$

is a *symmetric* tensor, which no longer contains partial derivatives of $\hat{\psi}$ with respect to the polarizations — i.e., it does not contain the local fields ${}^L\mathbf{E}_{\alpha}$ — and can thus be referred to as the “elastic stress” tensor because of its resemblance to the corresponding tensor of nonpolarized hyperelastic bodies. However, \hat{E}^t_{ij} still contains the polarizations via the functional dependence of $\hat{\psi}$. In particular, it contains the effects of electrostriction. Eq. (4.13) is the decomposition *a priori* set forth in Eq. (5.1) of Part One.

4.2. An alternate formulation

Instead of the objective reduced form (4.3) one may as well consider the following one:

$$(4.15) \quad \varrho_0 \psi = \Sigma = \bar{\Sigma}^{-1}(\mathbf{C}, \tilde{\Pi}_{\alpha}, \theta, \tilde{\mathbf{G}}), \quad \alpha = 1, 2, \dots, n,$$

where

$$(4.16) \quad \mathbf{C} \equiv \mathbf{F}^{-1}(\mathbf{F})^T, \quad \tilde{\Pi}_{\alpha} \equiv \varrho_0 \mathbf{F}^{-1} \pi_{\alpha} = J c_{\alpha}^{-1} \mathbf{F} \mathbf{P}_{\alpha},$$

$$\tilde{\mathbf{G}} \equiv \mathbf{F} \mathbf{g} = \mathbf{C} \mathbf{G}.$$

Then, in lieu of Eqs. (4.5), we have

$$(4.17) \quad \overset{-1}{\dot{C}}_{KL} = -2X_{K,i} D_{ij} X_{L,j}, \quad \overset{-1}{\dot{\Pi}}_{\alpha K} = J c_{\alpha}^{-1} X_{K,i} \dot{P}_{\alpha}.$$

The fact that D_{ij} and \dot{P}_{α} appear in these derivatives infers that the form (3.13) of the Clausius-Duhem inequality is more appropriate in the present formulation. In fact, we can rewrite the latter in the form

$$(4.18) \quad -(\dot{\Sigma} + \varrho_0 \eta \dot{\theta}) + J \hat{E}^t_{ij} D_{ij} - J \sum_{\alpha} {}^L\mathbf{E}_{\alpha} \cdot \dot{P}_{\alpha} - \theta^{-1} \tilde{\mathbf{Q}} \cdot \tilde{\mathbf{G}} \geq 0,$$

where

$$(4.19) \quad \tilde{\mathbf{Q}} \equiv J \mathbf{F}^T \tilde{\mathbf{q}} = J \mathbf{C} \hat{\mathbf{Q}}.$$

Computing $\dot{\Sigma}$ from Eq. (4.15) and using Eqs. (4.17), and assuming that the inequality (4.18) is to be satisfied for arbitrary independent objective time rates (D_{ij} , \dot{P}_{α} , $\dot{\theta}$, $\tilde{\mathbf{G}}$), we obtain the following results instead of Eqs. (4.7) through (4.11):

THEOREM. An alternate form of the constitutive equations of a nonmagnetized thermoelastic dielectric made of several molecular species is

$$(4.20) \quad \bar{F}^t_{ij} = -\frac{2}{J} \frac{\partial \bar{\Sigma}}{\partial C_{KL}} X_{K,i} X_{L,j} = \bar{E}^t_{ji},$$

$$(4.21) \quad {}^{RL}E_{\alpha i} = -c_{\alpha}^{-1} \frac{\partial \bar{\Sigma}}{\partial \tilde{\Pi}_{\alpha K}} X_{K,i}, \quad \alpha = 1, 2, \dots, n,$$

$$(4.22) \quad \eta = -\varrho_0^{-1} \frac{\partial \bar{\Sigma}}{\partial \theta}, \quad \Sigma = \bar{\Sigma}(\bar{\mathbf{C}}, \bar{\mathbf{\Pi}}_\alpha, \theta),$$

$$(4.23) \quad \tilde{q}_i = J^{-1} X_{X,i} \tilde{\mathbf{Q}}_\kappa(\bar{\mathbf{C}}, \bar{\mathbf{\Pi}}_\alpha, \theta, \tilde{\mathbf{G}}), \quad \tilde{\mathbf{Q}} \cdot \tilde{\mathbf{G}} \leq 0,$$

with

$$(4.24) \quad \tilde{\mathbf{Q}} = \mathbf{0}, \quad \text{whenever} \quad \tilde{\mathbf{G}} = \mathbf{0}.$$

Then the corresponding Cauchy stress follows from Eqs. (2.6) and (3.14) by eliminating σ_{ij} :

$$(4.25) \quad {}^R t_{ij} = E \bar{t}_{ij} + \varrho \sum_\alpha c_\alpha {}^{RL} E_{\alpha i} \pi_{\alpha j}$$

or, on account of Eqs. (4.20)–(4.21),

$$(4.26) \quad {}^R t_{ij} = -\frac{\varrho}{\varrho_0} \left(2 \frac{\partial \bar{\Sigma}}{\partial C_{KL}} X_{L,J} + J \sum_\alpha c_\alpha^{-1} \frac{\partial \bar{\Sigma}}{\partial \bar{\Pi}_{\alpha K}} P_{\alpha j} \right) X_{K,i}.$$

Both formulations (4.12)–(4.13) and (4.25)–(4.26) can be compared. Of course, on account of Eqs. (4.8) and (4.21), both equations (4.12) and (4.26) yield the moment-of-momentum balance law in the form

$$(4.27) \quad {}^R t_{[ij]} = \sum_\alpha {}^{RL} E_{\alpha [i} P_{\alpha j]},$$

which is the equation originally derived from the virtual power principle — cf. Eq. (4.14)₂ of Part One. The full correspondence between Eqs. (4.12) and (4.26) can be demonstrated by making a change of both potential function and independent variables, bearing in mind that the original functional dependence of ψ is that given by Eq. (4.2). We shall not give this proof here. However, in all cases, it must be remarked that ${}^R t_{ij}$ is not other than

$${}^R t_{ij} = \varrho \frac{\partial \psi}{\partial x_{i,\kappa}} x_{j,\kappa}$$

from which follows either Eq. (4.12) or Eq. (4.26), depending on whether one considers the reduced form (4.3) or (4.15) for the free energy density. Remark that each set of dependent variables present in Eq. (4.3) or (4.15) is consistent in the sense that each one uses either \mathbf{F} or $\bar{\mathbf{F}}$ to define the objective arguments, and not both at the same time. It is for that reason that Eqs. (4.12) and (4.26) take such forms as to allow one to recover directly Eq. (4.27) in both cases. The situation is much more involved if one considers a mixed set of independent variables (i.e., one variable constructed with \mathbf{F} and the other with $\bar{\mathbf{F}}$) as is the case if, specializing to the case of one polarization continuum and discarding polarization inertia,

$$(4.28) \quad \psi = \check{\psi}(\mathbf{E}, \bar{\mathbf{\Pi}}, \theta, \mathbf{G}).$$

Then one gets an equation of the type of Eq. (4.14) for ${}^E t_{ij}$ and an equation of the type of Eq. (4.21) for ${}^L \mathbf{E}$. However, according to Eq. (1–5.6), one then no longer needs to consider the Cauchy stress, the only stress tensor which remains in the formulation being the symmetric tensor ${}^E t_{ij}$. We consider this case from another standpoint in the next paragraph.

4.3. One polarization field and no polarization inertia

In this case the series of equations (2.9) is replaced by the single equation

$$(4.29) \quad \mathcal{E} + {}^L\mathbf{E} = \mathbf{0},$$

where ${}^L\mathbf{E}$ is the only local electric field appearing in the theory. As already remarked in Part One, the latter field can indeed be eliminated from the theory^(*) by Eq. (4.29). Performing this, one replaces Eqs. (2.6), (3.13), (3.14) and (4.25) by

$$(4.30) \quad t_{ij} = \sigma_{ij} - \mathcal{E}_{(i} P_{j)},$$

$$(4.31) \quad -\rho(\dot{\psi} + \eta\dot{\theta}) + {}^E t_{ij} D_{ij} + \mathcal{E} \cdot \dot{\mathbf{P}} - \theta^{-1} \tilde{\mathbf{q}} \cdot \nabla\theta \geq 0,$$

$$(4.32) \quad {}^E t_{ij} = \sigma_{ij} + \mathcal{E}_{(i} P_{j)},$$

$$(4.33) \quad {}^R t_{ij} = {}^E t_{ij} - \mathcal{E}_i P_j.$$

Then, according to Eq. (4.31), constitutive equations must in particular be constructed for ${}^E t_{ij}$ and \mathcal{E} . For hyperelastic solids one would consider a potential $\psi(\dots, \boldsymbol{\pi})$ to start with. However, one may now perform a Legendre transformation on ψ , so as to introduce a new free energy density F by

$$(4.34) \quad F(\dots, \mathcal{E}) = \psi(\dots, \boldsymbol{\pi}) - \mathcal{E} \cdot \boldsymbol{\pi}.$$

Noting that ψ and $\boldsymbol{\pi}$ are a scalar and a vectorial density, respectively, one then shows that Eq. (4.31) takes the form

$$(4.35) \quad -\rho(\dot{F} + \eta\dot{\theta}) + {}^E t_{ij} D_{ij} - \mathbf{P} \cdot \dot{\mathcal{E}} - \theta^{-1} \tilde{\mathbf{q}} \cdot \nabla\theta \geq 0$$

from which, in particular, will follow constitutive equations for ${}^E t_{ij}$ and \mathbf{P} . We do not compute these expressions. Rather, one may investigate the validity of the transformation performed starting with Eq. (4.29). Clearly, if ${}^L\mathbf{E}$ is a phenomenological field which describes *purely* thermodynamically reversible phenomena, then the transformation performed is legitimate. For this assertion to hold true it is also necessary that the entire field ${}^L\mathbf{E}$ be derivable from a potential. For this to be true it is necessary and sufficient that the orthogonal projection of ${}^L\mathbf{E}$ on $\dot{\mathbf{P}}$ be non-zero. Otherwise, one can always assume that one part of ${}^L\mathbf{E}$, noted ${}^L\mathbf{E}^\perp$, is such that

$$(4.36) \quad {}^L\mathbf{E}^\perp \cdot \dot{\mathbf{P}} = 0,$$

so that this part — which produces no power — would not be derivable from thermodynamical arguments. In conclusion, the transformations (4.30)–(4.35) are valid when: (i) only one polarization field is considered; (ii) the polarization inertia is negligible; (iii) both of the last two contributions of the following general decomposition vanish:

$$(4.37) \quad {}^L\mathbf{E} = {}^R\mathbf{L}\mathbf{E} + {}^D\mathbf{L}\mathbf{E} + {}^L\mathbf{E}^\perp.$$

Here, ${}^R\mathbf{L}\mathbf{E}$ is derivable from a potential, ${}^D\mathbf{L}\mathbf{E}$ is a dissipative contribution constructed with

^(*) See, however, the comment hereafter.

the help of a thermodynamical theory of irreversible processes, and ${}^L\mathbf{E}$, which satisfies Eq. (4.36), must be constructed in an *ad hoc* manner. As will be shown below, none of the conditions just enunciated are satisfied if one wants to account for the different optical effects in dielectrics.

4.4. Thermoelastic bodies with infinitesimal deformations

All constitutive equations derived in the foregoing sections can be linearized for small deformations and weak polarization field superimposed on a finite state of deformation (for the study of photomechanical effects) and polarization (for the study of electro-optical effects). In particular, the presence of several molecular species, hence of several polarization fields, enables us to reproduce the different states of dielectricity, e.g., *ferroelectricity*^(*) for which there exists a state of polarization in absence of applied electric field below the critical phase-transition temperature (in which case the choice of the polarization as an independent variable is clearly justified), and *antiferroelectricity* (for which two antiparallel polarization fields of equal magnitude in absence of externally applied field are necessary; cf. [7]), as well as the most general case of *ferrielectricity*. This linearization will be performed in a subsequent paper devoted to wave propagation in such media. For the time being, we consider only a linearization for the case of infinitesimal deformations and small temperature variations about an initial strain-free state with spatially uniform temperature $\theta^{0(**)}$. In these conditions, noting u the displacement field and setting $\tilde{\theta} \equiv \theta - \theta^0$, we write (cf. [9] Chap. 6):

$$(4.38) \quad \begin{aligned} x_{i,K} &\simeq (\delta_{ij} + u_{i,j})\delta_{jK}, & E_{KL} &\simeq e_{ij}\delta_{iK}\delta_{jL}, & e_{ij} &\equiv u_{(i,j)}, \\ \rho &\simeq \rho_0(1 - e_{kk}), & J &\simeq 1 \end{aligned}$$

and, from either one of the formulations given in Sects. 4.1 and 4.2, we obtain

$$(4.39) \quad \begin{aligned} E_{t_{ij}} &= \rho_0 \frac{\partial \psi}{\partial e_{ij}}, & {}^{RL}\mathbf{E}_\alpha &= -c_\alpha^{-1} \frac{\partial \psi}{\partial \pi_\alpha}, & \eta &= -\frac{\partial \psi}{\partial \tilde{\theta}}, \\ {}^{RL}t_{ij} &= E_{t_{ij}} - \rho_0 \sum_\alpha c_\alpha \pi_{\alpha i} {}^{RL}E_{\alpha j}. \end{aligned}$$

Equations (4.38) are valid as δ goes to zero, δ being an infinitesimally small of the first order such that $|\nabla u| < \delta$. As regards heat flux, it is of course assumed that $|g| < \delta_h$, where δ_h is infinitesimally small. On account of the continuity condition (4.11) or (4.24), a Taylor series expansion about $\nabla\theta = \mathbf{0}$ yields the classical Fourier law

$$(4.40) \quad \tilde{q}_i = -K_{ij}(\theta^0)\theta_{,j} + o(\delta_h)$$

as δ_h goes to zero. Of course, $K_{ij} = K_{ji}$, and $K_{ij}a_i a_j \geq 0$ for any \mathbf{a} . In writing the functional dependence of K_{ij} , we have discarded the coupling of heat conduction with the thermodynamically recoverable phenomena.

(*) However, one then has probably to consider the spatial nonuniformities, hence the gradients of polarization, to account for the ordering effect in a continuous manner (compare the ferromagnetic and ferrimagnetic cases in Refs. [6], [8]).

(**) An essential difference with the case of the linearization about bias fields is that only the strain tensor, i.e., the symmetric part of the displacement gradient, is involved here.

It remains to give the expression for the free energy. For the theory of infinitesimal deformations, the following expansion in e_{ij} and $\tilde{\theta}$ seems reasonable

$$(4.41) \quad \psi = \frac{\Sigma}{\varrho_0} = \psi_0 - \eta_0 \tilde{\theta} - \frac{C}{2\varrho_0} \tilde{\theta}^2 - \varrho_0^{-1} \tilde{\theta} \nu_{ij}(\theta^0) e_{ij} - \tilde{\theta} \sum_{\alpha} c_{\alpha} a_i^{(\alpha)}(\theta^0) \pi_{\alpha i} \\ + \frac{1}{2\varrho_0} \lambda_{ijkl}(\theta^0) e_{ij} e_{kl} + \sum_{\alpha} c_{\alpha} f_{ijk}^{(\alpha)}(\theta^0) e_{ij} \pi_{\alpha k} + \varrho_0 \sum_{\alpha, \beta} \frac{1}{2} c_{\alpha} c_{\beta} \gamma_{ijkl}^{(\alpha\beta)}(\theta^0) e_{ij} \pi_{\alpha k} \pi_{\beta l} \\ + \varrho_0 \sum_{\alpha, \beta} \frac{1}{2} c_{\alpha} c_{\beta} a_{ij}^{(\alpha\beta)}(\theta^0) \pi_{\alpha i} \pi_{\beta j} + \text{h.o.t. in } \pi_{\alpha} \otimes \pi_{\beta}.$$

The factors ϱ_0 and c_{α} are introduced for convenience. The tensor coefficients introduced satisfy obvious symmetry relations that we do not reproduce here, but the number of their independent components is still very large. This number can be drastically reduced if we remark the following. The temperature has practically the same effect on each polarization field or *sub-lattice polarization* (to conform more closely to the physical terminology) π_{α} . Thus, $\mathbf{a}^{(\alpha)}$ does not depend on α . Calling $\boldsymbol{\pi}$ the total polarization per unit mass, the fifth term in Eq. (4.41) yields $-\tilde{\theta} \mathbf{a}(\theta^0) \cdot \boldsymbol{\pi}$. Also, the interactions between the material lattice (substrata of elastic deformations) and each sub-lattice polarization π_{α} assumes the same form. Hence, $f_{ijk}^{(\alpha)}$ does not depend on α . The interactions within each polarization sub-lattice are formally the same; thus $\gamma_{ijkl}^{(\alpha\beta)}$ does not depend on α , and similarly for $a_{ij}^{(\alpha\beta)}$. The interactions within each couple of differing polarization sub-lattices are assumed to have the same form. This is so much the better because the individual sub-lattice polarizations are macroscopically unobservable quantities (only the total polarization is observable), so that the material coefficients labeled α or β could not be measurable. On account of these remarks, Eq. (4.41) can be rewritten as

$$(4.42) \quad \psi = \frac{\Sigma}{\varrho_0} = \psi_0 - \eta_0 \tilde{\theta} - \frac{C}{2\varrho_0} \tilde{\theta}^2 - \varrho_0^{-1} \tilde{\theta} \nu_{ij} e_{ij} - \tilde{\theta} \mathbf{a} \cdot \boldsymbol{\pi} + \frac{1}{2\varrho_0} \lambda_{ijkl} e_{ij} e_{kl} \\ + f_{ijk} e_{ij} \pi_k + \frac{1}{2} \varrho_0 \gamma_{ijkl}^{(ee)} e_{ij} \left(\sum_{\alpha} c_{\alpha}^2 \pi_{\alpha k} \pi_{\alpha l} \right) + \varrho_0 \gamma_{ijkl}^{(ee)} e_{ij} \left(\sum_{\alpha \neq \beta} c_{\alpha} c_{\beta} \pi_{\alpha k} \pi_{\beta l} \right) \\ + \frac{1}{2} \varrho_0 a_{ij} \left(\sum_{\alpha} c_{\alpha}^2 \pi_{\alpha i} \pi_{\alpha j} \right) + \varrho_0 b_{ij} \left(\sum_{\alpha \neq \beta} c_{\alpha} c_{\beta} \pi_{\alpha i} \pi_{\beta j} \right) + \text{h.o.t.}$$

on account of Eq. (2.8)₁^(*). The remaining tensor coefficients satisfy obvious symmetry relations.

(*) For instance, for isotropy and limiting the description to two polarization sub-lattices ($\alpha = 1, 2$), the last two terms in Eq. (4.42) yield a volume energy

$$\Sigma = \frac{a}{2} (\mathbf{P}_1^2 + \mathbf{P}_2^2) + b \mathbf{P}_1 \cdot \mathbf{P}_2 + \text{h.o.t.}$$

which can be compared with the expression considered by KITTEL [10] in his phenomenological approach to rigid antiferroelectrics.

On account of Eq. (4.42), Eq. (4.39) yields the following constitutive equations:

$$(4.43) \quad \eta = \eta_0 + \frac{C}{\varrho_0} \tilde{\theta} + \varrho_0^{-1} \nu_{ij} e_{ij} + \mathbf{a} \cdot \boldsymbol{\pi},$$

$$(4.44) \quad {}^E t_{ij} = -\tilde{\theta} \nu_{ij} + \lambda_{ijkl} e_{kl} + f_{ijk} P_k + \frac{1}{2} \gamma_{ijkl}^{(e)} \left(\sum_{\alpha} P_{\alpha k} P_{\alpha l} \right) + \gamma_{ijkl}^{(ee)} \left(\sum_{\alpha \neq \beta} P_{\alpha k} P_{\beta l} \right),$$

$$(4.45) \quad {}^{RL} E_{\alpha i} = \tilde{\theta} a_i - e_{pq} [f_{pqi} + \gamma_{pqik}^{(e)} P_{\alpha k} + \gamma_{pqik}^{(ee)} (P_k - P_{\alpha k})] - a_{ij} P_{\alpha j} - b_{ij} (P_j - P_{\alpha j}),$$

$$(4.46) \quad {}^R t_{ij} = -\tilde{\theta} (\nu_{ij} + P_i a_j) + \lambda_{ijkl} e_{kl} + (f_{ijk} P_k + f_{pqj} e_{pq} P_i) \\ + \sum_{\alpha} \left(\frac{1}{2} \gamma_{ijkl}^{(e)} P_{\alpha k} P_{\alpha l} + \gamma_{pqjk}^{(e)} e_{pq} P_{\alpha k} P_{\alpha i} \right) + \gamma_{ijkl}^{(ee)} \left(\sum_{\alpha \neq \beta} P_{\alpha k} P_{\beta l} \right) \\ + \gamma_{pqjk}^{(ee)} e_{pq} \sum_{\alpha} P_{\alpha i} (P_k - P_{\alpha k}) + a_{jk} \left(\sum_{\alpha} P_{\alpha i} P_{\alpha k} \right) + b_{jk} \sum_{\alpha} P_{\alpha i} (P_k - P_{\alpha k}),$$

on account of the fact that

$$\varrho_0 \sum_{\beta \neq \alpha} c_{\beta} \boldsymbol{\pi}_{\beta} = \mathbf{P} - \mathbf{P}_{\alpha}.$$

In the above equations (4.42)–(4.46), the different material coefficients introduced are easily identified as: C = heat capacity; ν_{ij} = thermoelasticity tensor; a_i = pyroelectricity vector; λ_{ijkl} = elasticity moduli; f_{ijk} = piezoelectricity tensor; $\gamma_{ijkl}^{(e)}$ and $\gamma_{ijkl}^{(ee)}$ = electrostrictive tensors; a_{ij} = “dielectric” tensor; b_{ij} = inter-polarization sub-lattice interactions. The coefficients $\gamma_{ijkl}^{(ee)}$ and b_{ij} only are proper to the theory accounting for several molecular species. The constitutive equations which follow are, of course, linear in the strains, but they are quadratic in the polarizations. Furthermore, if we are close to a transition point, approaching it from below (e.g. in ferroelectrics) then, because of the critical behavior of the polarizations at that point, one should consider some higher order terms in $\boldsymbol{\pi}_{\alpha} \otimes \boldsymbol{\pi}_{\beta}$ in the expansions (4.41) and (4.42), rising critically the order of Eqs. (4.43)–(4.46) in $\boldsymbol{\pi}_{\alpha}$ by the same token. As indicated when introducing the tensor ${}^E t_{ij}$ — cf. comments after Eq. (4.14) — the equation (4.44) accounts for the “elastic” effects: thermoelasticity, piezoelectricity and electrostriction, whereas the Cauchy stress (4.46), which is not symmetric, accounts for all interactions (for instance, the pyroelectric effect and the interactions between differing polarization sub-lattices participate in ${}^E t_{ij}$).

Simple approximations can be deduced from Eqs. (4.44)–(4.46). First, one may consider constitutive equations which are jointly linear in e_{ij} and $P_{\alpha i}$ (thus neglecting terms in $e \otimes \boldsymbol{\pi}_{\alpha}$, $\boldsymbol{\pi}_{\alpha} \otimes \boldsymbol{\pi}_{\beta}$, and higher order terms). Then, in particular, electrostrictive effects are discarded. We have thus

$$(4.47) \quad {}^R t_{ij} = {}^E t_{ij} = -\tilde{\theta} \nu_{ij} + \lambda_{ijkl} e_{kl} + f_{ijk} P_k,$$

$$(4.48) \quad {}^{RL} E_{\alpha i} = \tilde{\theta} a_i - f_{pqi} e_{pq} - a_{ij} P_{\alpha j} - b_{ij} (P_j - P_{\alpha j}).$$

The fact that ${}^R t_{ij}$ is here reduced to the symmetric tensor ${}^E t_{ij}$ means that the local balance law of angular momentum — cf. Eq. 1–(4.14)₂ — is no longer satisfied in this approximation, unless we neglect the ponderomotive couple.

The standard theory of piezoelectricity [11]–[12] is recovered as follows. First, thermoelastic and pyroelectric effects are neglected, i.e., $\nu_{ij} = 0$, $a_i = 0$. Next, only one polarization is considered. Finally, the only remaining equation (2.9), written in electrostatics and in which the inertia is supposed to be negligibly small, yields

$$(4.49) \quad \mathbf{E} + {}^{RL}\mathbf{E} = \mathbf{0}.$$

On account of Eq. (4.48) and of the different assumptions made, Eqs. (4.47) and (4.49) yield

$$(4.50) \quad \begin{aligned} {}^E t_{ij} &= \lambda_{ijkl} e_{kl} + f_{ijk} P_k, \\ {}^E E_i &= f_{pq} e_{pq} + a_{ij} P_j. \end{aligned}$$

These are VOIGT's [13] constitutive equations of piezoelectricity expressed with the independent variables e_{ij} and P_i .

5. Voigt's intramolecular force balance in elastic dielectrics

The linearization with respect to infinitesimal deformations performed in Sect. 4.4 being assumed, we can consider, to illustrate the influence of loss mechanisms, that thermodynamically recoverable contributions (already labeled R) and irreversible contributions (from hereon labeled D) are additive in the different constitutive equations. That is, given the linearity with respect to the dependent constitutive variables of the decompositions (2.6), (3.14), (4.13) and (4.25), and taking account of a general decomposition of the type (4.37) for each ${}^L E_\alpha$, we can write

$$(5.1) \quad \begin{aligned} t_{ij} &= {}^D t_{ij} + {}^R t_{ij}, \\ {}^R t_{ij} &= {}^E t_{ij} - \varrho_0 \sum_{\alpha} c_{\alpha} \pi_{\alpha i} {}^{RL} E_{\alpha j}, \\ {}^D t_{ij} &= {}^V t_{ij} - \varrho_0 \sum_{\alpha} c_{\alpha} \pi_{\alpha i} {}^{DL} E_{\alpha j}, \\ {}^L E_{\alpha} &= {}^{RL} E_{\alpha} + {}^{DL} E_{\alpha} + {}^{\perp} E_{\alpha}, \end{aligned}$$

where, according to the formulation (4.18),

$$\varrho \dot{\psi} = -\varrho \eta \dot{\theta} + {}^E t_{ij} D_{ij} - \sum_{\alpha} {}^{RL} E_{\alpha} \cdot \dot{\mathbf{P}}_{\alpha},$$

whereas ${}^V t_{ij}$, the viscous stress tensor (the analogue of ${}^E t_{ij}$ for dissipative processes), ${}^{DL} E_{\alpha}$ and $\tilde{\mathbf{q}}$ satisfy the remaining dissipation inequality

$$(5.2) \quad \Phi \equiv {}^V t_{ij} D_{ij} - \sum_{\alpha} {}^{DL} E_{\alpha} \cdot \dot{\mathbf{P}}_{\alpha} - \theta^{-1} \tilde{\mathbf{q}} \cdot \nabla \theta \geq 0,$$

and the ${}^{\perp} E_{\alpha}$'s satisfy the orthogonality conditions:

$$(5.3) \quad {}^{\perp} E_{\alpha} \cdot \dot{\mathbf{P}}_{\alpha} = 0, \quad \alpha = 1, 2, \dots, n.$$

On account of the assumed linearization for the constitutive equations representing the reversible processes, we can use the Onsager-Casimir theory to express the variables v_{ij} , ${}^D L E_\alpha$ and \tilde{q} , and similarly assume a linear expression for each ${}^L \dot{E}_\alpha$. Furthermore, we assume that each contribution in Eq. (5.2) is disconnected from the other ones. Hence, introducing the phenomenological tensorial coefficients η_{ijkl} , $c_{ij}^{(\alpha\beta)}$, K_{ij} and $G_{ij}^{(\alpha)}$ such that

$$(5.4) \quad \eta_{ijkl} D_{ij} D_{kl} \geq 0, \quad \sum_{\alpha, \beta} c_{ij}^{(\alpha\beta)} \dot{P}_{\alpha i} \dot{P}_{\beta j} \geq 0,$$

$$K_{ij} g_i g_j \geq 0, \quad G_{ij}^{(\alpha)} \dot{P}_{\alpha i} \dot{P}_{\alpha j} = 0,$$

we can write

$$(5.5) \quad v_{ij} = \eta_{ijkl} D_{kl}, \quad {}^D L E_{\alpha i} = - \sum_{\beta} c_{ij}^{(\alpha\beta)} \dot{P}_{\beta j},$$

$$\tilde{q}_i = -K_{ij} \theta_{,j}, \quad {}^L \dot{E}_{\alpha i} = -G_{ij}^{(\alpha)} \dot{P}_{\alpha j}.$$

The last of Eqs. (5.4) requires that $G_{ij}^{(\alpha)}$ be a skew-symmetric tensor, so that we can introduce its dual axial vector $G_i^{(\alpha)}$ and rewrite the last of Eqs. (5.5) as

$$(5.6) \quad {}^L \dot{E}_\alpha = \dot{\mathbf{G}}^{(\alpha)} \times \mathbf{P}_\alpha.$$

There are no axial vectors in the present theory of unmagnetized dielectrics except the magnetic induction \mathbf{B} . Furthermore, to obtain a linear equation in time-varying fields in Eq. (5.6), only the static part B_0 of B , if it exists, can be considered. Introducing the absolute scalar coefficients Γ_α , Eq. (5.6) can be rewritten as

$$(5.7) \quad {}^L \dot{E}_\alpha = \Gamma_\alpha \mathbf{B}_0 \times \dot{\mathbf{P}}_\alpha.$$

Finally, Eqs. (5.5)₁₋₂ and (5.1)₃ provide the dissipative part of the Cauchy stress in the form

$$(5.8) \quad {}^D t_{ij} = \eta_{ijkl} D_{kl} + \sum_{\alpha, \beta} P_{\alpha i} c_{jk}^{(\alpha\beta)} \dot{P}_{\beta k}.$$

If a linearization is performed for studying the propagation of small perturbations, then $D_{ij} = \partial e_{ij} / \partial t = \dot{e}_{ij}$, whereas the second contribution in Eq. (5.8) will be zero except if there is some initial static polarization; such a polarization will exist in a classical dielectric if there is a nonzero electric bias field. However, it will also exist in ferroelectrics and ferrielectrics even if the static electric field is zero. Thus we have a result similar to that obtained in deformable ferromagnets [5]: the loss mechanisms associated with the dielectric effects in general contribute to the Cauchy stress. This shows, without studying any specific wave propagation problem, that these loss mechanisms will participate, along with the usual viscosity processes, in the damping of elastic waves if there is some initial polarization.

There is no difficulty to write down the complete expression (5.1)₁ of the Cauchy stress on account of Eq. (5.8) and the results of the previous sections. More interesting

for the present purpose is the complete expression of Eq. (2.9). On account of Eqs. (5.1)_s, (5.5)₂ and (5.7), Eq. (2.9) yields

$$(5.9) \quad \mathcal{E}_i + {}^{RL}E_{ai} - \sum_{\beta} c_{ij}^{(\alpha\beta)} \dot{P}_{\beta j} + \Gamma_{\alpha} (B_0 \times \dot{P}_{\alpha})_i = d_{\alpha} \ddot{\pi}_i.$$

For the case of infinitesimal deformations, denoting by \mathbf{E}_0 an initial static electric field and by \mathbf{e} a perturbation, so that Eq. (2.5) gives

$$(5.10) \quad \mathcal{E} = \mathbf{E}_0 + \mathbf{e} + \frac{1}{c} \dot{\mathbf{u}} \times \mathbf{B}_0,$$

noting also that, $\mathbf{P}_{\alpha}^{(0)}$ being the initial static polarizations,

$$(5.11) \quad \dot{P}_{\alpha} \approx \frac{\partial P_{\alpha}^{(a)}}{\partial t} - (\mathbf{P}_{\alpha}^{(0)} \cdot \nabla) \frac{\partial u}{\partial t} + \mathbf{P}_{\alpha}^{(0)} \left(\nabla \cdot \frac{\partial u}{\partial t} \right) \equiv \frac{\tilde{\partial} P_{\alpha}}{\partial t},$$

$$d_{\alpha} \ddot{\pi}_{\alpha} \approx a_{\alpha} \frac{\partial^2 P_{\alpha}}{\partial t^2} \quad (a_{\alpha} \equiv d_{\alpha} / \epsilon_0 c_{\alpha}),$$

and taking account of Eq. (4.45), Eq. (5.9) gives for $\alpha = 1, 2, \dots, n$:

$$(5.12) \quad E_{0i} + e_i = a_{\alpha} \frac{\partial^2 P_{\alpha i}}{\partial t^2} + \sum_{\beta=1}^n c_{ij}^{(\alpha\beta)} \frac{\tilde{\partial} P_{\beta j}}{\partial t} + \left[\left(\Gamma_{\alpha} \frac{\tilde{\partial} P_{\alpha}}{\partial t} - \frac{1}{c} \frac{\partial u}{\partial t} \right) \times B_0 \right]_i + a_{ij} P_{\alpha j}$$

$$+ b_{ij} (P_j - P_{\alpha j}) - \theta a_i + e_{pq} [f_{pqi} + \gamma_{pqik}^{(e)} P_{\alpha k} + \gamma_{pqik}^{(ee)} (P_k - P_{\alpha k})].$$

If we compare this equation to the following equation proposed by VOIGT [14] in his pioneering work devoted to rigid stationary isotropic dielectrics^(*):

$$(5.13) \quad \mathbf{P}_{\alpha} + \bar{a}_{\alpha} \frac{\partial^2 \mathbf{P}_{\alpha}}{\partial t^2} + \bar{b}_{\alpha} \frac{\partial \mathbf{P}_{\alpha}}{\partial t} + \bar{\Gamma}_{\alpha} \frac{\partial \mathbf{P}_{\alpha}}{\partial t} \times \mathbf{B}_0 = \bar{k}_{\alpha} \mathbf{E},$$

we may claim that Eqs. (5.12) are the generalization of Voigt's (5.13) *intramolecular force balance equations* to the case of elastic anisotropic dielectrics made of several polarization sub-lattices, by accounting for the static and dynamical electromagnetic fields, the effects of pyroelectricity, piezoelectricity and electrostriction, and the coupling between the different polarization sub-lattices and the associated loss mechanisms. Furthermore, Eqs. (5.12) are valid for all states of dielectricity (e.g., ferroelectricity, and antiferroelectricity). For isotropy in a rigid stationary body, Eq. (5.12), however, does not reduce exactly to Eq. (5.13), since we obtain

$$(5.14) \quad \mathbf{E} = a_{\alpha} \frac{\partial^2 \mathbf{P}_{\alpha}}{\partial t^2} + c^{(\alpha\alpha)} \frac{\partial \mathbf{P}_{\alpha}}{\partial t} + \Gamma_{\alpha} \frac{\partial \mathbf{P}_{\alpha}}{\partial t} \times B_0 + a \mathbf{P}_{\alpha} + b (\mathbf{P} - \mathbf{P}_{\alpha}) + c^{(\alpha\beta)} \frac{\partial}{\partial t} (\mathbf{P} - \mathbf{P}_{\alpha}),$$

the last two terms arising from the interactions between the different polarization sub-lattices (we have assumed that $c^{(\alpha\beta)}$ is independent of the choice of the couple $\alpha \neq \beta$). Eq. (5.13) is recovered only if $b = 0$ and $c^{(\alpha\beta)} = 0$ for $\alpha \neq \beta$, i.e., if these interactions are discarded.

^(*) The coefficients in Eqs. (5.12) and (5.13) do not have the same values.

Some remarks should be made concerning equations of the type (5.12) or (5.13). First, it must be remarked that in taking the time-derivatives of the polarizations in Eqs. (5.12), (5.13) and (5.14), we need to consider only the time-varying perturbation part of the \mathbf{P}_α 's, i.e.

$$(5.15) \quad \mathbf{p}_\alpha \equiv \mathbf{P}_\alpha - \mathbf{P}_\alpha^{(0)}.$$

Next, the equations (5.12) and (5.13) are true *balance laws* in which constitutive equations have been carried for ${}^L E_\alpha$, and are not constitutive equations of the rate-type (up to second time derivatives) as it has been advanced by certain authors (e.g., in [15]). In particular, the second time-derivative belongs to a real "inertia" term, as it was demonstrated in Part One, and suspected in the early work of TOUPIN [16]. In fact, Eq. (5.12) or, simpler, the equations (5.13) and (5.14) — which are not coupled with the deformation field — is, apart from the term involving Γ_α or $\bar{\Gamma}_\alpha$, closely related to the *resonance equation* built after a simple mechanical model and used to study, in an elementary fashion, the damping and relaxation phenomena in rigid dielectrics (cf., for instance, [7] Eq. 16.4; also [17]–[18]). Hence Eq. (5.12), when supplemented with Cauchy's equation and Maxwell's equations, serves to study the same phenomena coupled with elastic deformations. Finally, terms of the type of those that contain the constant Γ_α or $\bar{\Gamma}_\alpha$ are known to provide an *ad hoc* description of the Faraday effect in dielectrics (cf. [14], [16]). It has been argued [19] that the introduction of such terms was not "rational" because they include the magnetic induction whereas all other constitutive equations are supposed not to depend on \mathbf{B} , so that the so-called "principle" of equipresence would be violated. We recall that "equipresence" is only a precautionary measure^(*). Anyhow, one can always suppose that all dependent constitutive arguments depend on \mathbf{B} to start with. Then, using the Clausius-Duhem inequality in which there appears no time rate $\dot{\mathbf{B}}$, all constitutive equations will be shown to be independent of \mathbf{B} , except ${}^L E_\alpha$ which, after Eq. (5.3), does not contribute to this inequality, so that nothing can be decided concerning this field for which the approximation (5.7) seems satisfactory in the linear theory. We thus conclude that there is no need to consider weakly magnetizable viscoelastic dielectrics to substantiate the Faraday effect. Finally, the above equations can be related to other simpler equations found in the literature. For instance, for a rigid anisotropic dielectric, considering only one polarization field, in absence of static magnetic induction, and if the polarization inertia is negligible (which is most often the case), Eq. (5.12) reduces to

$$(5.16) \quad E_i = a_{ij} P_j + c_{ij} \frac{\partial P_j}{\partial t}.$$

This equation, which is a generalization of the Maxwell-Neumann equation of classical photoelasticity (if a_{ij} and c_{ij} depend on the strains), and which describes a birefringent gyrotropic crystal with dichroism^(**), is given by RAMACHANDRAN and RAMASHESHAN [21]. The presence of the dissipative contribution $c_{ij} \partial P_j / \partial t$ indicates that such a crystal in general is not transparent, i.e., it produces an absorption of electromagnetic waves.

^(*) As was already noted by several authors, it is in fact impossible to call a "principle" something that is regularly violated (e.g., the free energy does not depend on the temperature gradient at the output).

^(**) See the treatise of BORN [20] for these notions.

In conclusion of these remarks, we want to emphasize the evident wealth and the rich possibilities offered by Eq. (5.12) and the allied Cauchy's equations for studying coupled electromechanical and electro-optical effects in elastic dielectrics, ferroelectrics and anti-ferroelectrics. This must be the concern of further researches. In particular, the treatments of TOKUOKA and KOBAYASHI [22]–[23] could be extended to the case of elastic ferroelectrics.

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UNIVERSITÉ DE PARIS VI,
LABORATOIRE DE MÉCANIQUE THÉORIQUE ASSOCIÉ AU C.N.R.S.

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